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Abstract. Highest atmospheric ozone production rates can be found at around 30 km in the tropical stratosphere, leading to ozone mixing ratios of about 10 ppmv. Those stratospheric air masses are then transported to extra-tropical latitudes via the Brewer-Dobson circulation. This is considered the main mechanism to generate mid- and high latitude ozone. By applying the climate-chemistry models E39/C and MAECHAM4/CHEM, this view is investigated in more detail. The origin of ozone in the troposphere and stratosphere is analysed, by incorporating a diagnostics ("marked ozone origin tracers") into the models, which allows to identify the origin of ozone. In most regions the simulated local ozone concentration is dominated by local ozone production, i.e. less than 50% of the ozone at higher latitudes of the stratosphere is produced in the tropics, which conflicts with the idea that the tropics are the global source for stratospheric ozone. Although episodic stratospheric intrusions occur basically everywhere, the main ozone stratosphere-to-troposphere exchange is connected to exchange processes at the sub-tropical jet-stream. The simulated tropospheric influx of ozone amounts to 420 Tg per year, and originates in the Northern Hemisphere from the extra-tropical stratosphere, whereas in the Southern Hemisphere a re-circulation of tropical tropospheric ozone contributes most to the influx of ozone into the troposphere. In the model E39/C, the upper troposphere of both hemispheres is clearly dominated by tropical tropospheric ozone (40%–50%) except for northern summer hemisphere, where the tropospheric contribution (from the tropics as well as from the Northern Hemisphere) does not exceed 20%.

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1 Introduction

The ozone distribution in the lower stratosphere is generally considered to be controlled by tropical ozone production and, below 30 km, by large-scale transport, i.e. the Brewer-Dobson circulation (BDC), especially in winter (World Meteorological Organization (WMO), 1999). Air masses are uplifted in the tropics, then transported to the winter pole and subside during winter. However, transport time-scales and the stratospheric age of air exceeds two years (Hall and Plumb, 1994) meaning that the seasonal cycle has to be taken into account. Early studies of the meridional circulation (Dunkerton, 1978) in a Lagrangian view suggest that the air masses are rising in the tropics, transported to higher latitudes, but then are swapped northward and southward according to the direction of the BDC during the course of year. This makes the distinction between dynamical and chemical implications for ozone more complicated, since the chemical life-time of ozone highly depends on the time of year and on latitude. Furthermore, it has to be noted that wave-breaking in the lower-most stratosphere results in transport of tropical air masses to higher latitudes through the sub-tropical barrier even at lower stratospheric altitudes. These so-called streamers were frequently observed (e.g. Offermann et al., 1999) and transport low ozone air masses to higher latitudes in all seasons, depending on strength of the storm tracks (Eyring et al., 2003). It has been speculated that this process may have an important contribution to the mid-latitude ozone concentration at the tropopause altitudes (Grewe et al., 2004). The WMO report (1999) summarises this by pointing out that the classical view of the BDC has been significantly refined in recent years, which has consequences on the ozone controlling processes.

This paper aims at challenging the view that the tropical stratospheric region is the dominant source of ozone, at least at higher latitudes and investigates its implications on stratosphere-troposphere exchange and tropospheric ozone.

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Table 1. Areas of interest for the origin of ozone. NH, TR, and SH describe Northern Hemisphere, Tropics, and Southern Hemisphere. TS, LS, and MS denote troposphere, lower stratosphere, and middle stratosphere. The two values for the upper boundaries of the MS region refer to the respective models.

	Latitude	Pressure [hPa]
NHTS	90°N–30°N	1000–200
TRTS	30°N–30°S	1000–100
SHTS	30°S–90°S	1000–200
NHLS	90°N–30°N	200–100
TRLS	30°N–30°S	100–40
SHLS	30°S–90°S	200–100
NHMS	90°N–30°N	100–10/0.01
TRMS	30°N–30°S	40–10/0.01
SHMS	30°S–90°S	100–10/0.01

The applied methodology is based on the separation of the contribution of large-scale transport from the contribution of chemistry to local ozone concentrations for distinct regions. The varying importance of dynamics and chemistry during the course of the year is taken into account and plays a major role. The basic question to be answered is: Taking the ozone concentration at any given location, in which regions has this ozone been produced? This has an implication on estimates of trends, because, if we know the contributions from different regions to a local ozone concentration, we can estimate the impact of variations or trends in large-scale transport on trends in ozone.

To answer those questions the fully coupled climate-chemistry model E39/C is applied, which includes both troposphere and stratosphere dynamics and chemistry. A summary of publications dedicated to validate E39/C in terms of mean state and processes is given in Sect. 3. This model has its upper level centered at 10 hPa, which somehow limits its representation of stratospheric dynamics. To better understand these limitation for the objective of this paper the middle atmosphere version of ECHAM4/CHEM, i.e. MAECHAM4/CHEM, has been applied and compared to the results of E39/C. In both models a number of diagnostic ozone tracers are included, which label ozone molecules according to their production region. The models and the diagnostic technique are described in Sects. 2 and 4. Section 5 addresses the origin of stratospheric ozone, whereas Sect. 6 describes the origin of ozone transported into the troposphere. Section 7 discussed the production and transport of tropospheric ozone. In Sect. 8, a comparison to observational data is provided including a discussion of the implication on the findings of the preceding sections.

2 Model description

For the determination of the origin of ozone, the coupled chemistry-climate model E39/C is applied, which consists of the spectral atmospheric general circulation model ECHAM4.L39(DLR) (E39) and the chemistry module CHEM (C). E39 is based on the climate model ECHAM4 (Roeckner et al., 1996) with increased vertical resolution from 19 to 39 levels and the top layer centered at 10 hPa (Land et al., 1999). In this study, the model was applied with a horizontal resolution of T30, i.e. a corresponding grid size of $3.75° \times 3.75°$. Water vapour, cloud water and chemical species are advected by a so-called semi-Lagrangian scheme. E39 includes parameterisation schemes for small scale physical processes, like convection or cloud formation. Convection is parameterised with a mass-flux scheme (Tiedtke, 1989). Cumulus clouds are represented by a bulk model including the effect of entrainment and detrainment on the updraft and downdraft of convective mass fluxes. Three types of convection are distinguished: Tropical deep convection (low level synoptic scale convergence); shallow cumulus convection, e.g. tradewind cumuli; mid-level convection representing the convective cells, which have their roots not in the boundary layer but originate at levels above (more details can be found in e.g. Brinkop, 2002).

The model has been used in a variety of tracer transport studies (Land, 1999; Land et al., 2002; Rogers et al., 2002; Timmreck et al., 1999), including e.g., ^{222}Rn experiments to evaluate fast vertical transport from the boundary layer to upper levels, which largely can be associated to convective transport. The chemistry module CHEM (Steil et al., 1998) is based on the family concept. It contains the most relevant chemical processes for describing the tropospheric background NO_x-CH_4-CO-HO_x-O_3 chemistry as well as the stratospheric homogeneous and heterogeneous ozone chemistry. CHEM includes 37 chemical species of which 9 are explicitly transported and the others are grouped into 3 families. 107 photochemical reactions and 4 heterogeneous reactions on polar stratospheric clouds (PSCs) and on sulphate aerosols are considered in CHEM, however not yet bromine chemistry. Mixing ratios of methane (CH_4), nitrous oxide (N_2O) and carbon monoxide (CO) are prescribed at the surface. Nitrogen oxide emissions at the surface (natural and anthropogenic sources) and from aircraft are considered. Lightning NO_x emissions are calculated interactively depending on the mass fluxes in deep convective clouds according to Grewe et al. (2001).

At the uppermost level of E39/C total NO_y and total Cl_y are prescribed at every timestep including a seasonal cycle and latitudinal dependencies, derived from a 2-D model (further details are given in Hein et al., 2001). For all tracers (including ozone) a zero vertical flux is assumed at the top of the model.

For a better understanding of the impact of the upper boundary conditions applied in E39/C on the stratospheric

results, the middle atmosphere model MAECHAM4/CHEM (Steil et al., 2003) has been employed in addition. It consists of the middle atmosphere ECHAM4 version (Manzini et al., 1997; Manzini and McFarlane, 1998) and basically the same chemical module CHEM as used in E39/C, with additionally explicit transport and chemistry of CFCs.

Model climatologies and detailed descriptions are given in Hein et al. (2001) and Grewe et al. (2001) for E39/C and in Steil et al. (2003), Manzini and Feichter (1999) and Manzini et al. (2003) for MAECHAM4/CHEM. The model E39/C has been applied for various chemistry-transport and climate-chemistry investigations (Schnadt et al., 2002; Grewe et al., 2002; Dameris et al., 2005) and contributed to model intercomparisons (e.g., Austin et al., 2003). Recently, Dameris et al. (2005, 2006) showed that ozone variations during the last 40 years can be reproduced in agreement with observations by employing realistic external forcings (e.g. volcanic eruptions, El Niño, Quasi-Biennial Oscillation, 11-year solar cycle). A detailed overview on the models capabilities and disabilities is given in the next section

For this investigation 5 year runs (excluding a spin-up time) have been performed for both models applying constant boundary conditions for 1990 and including additionally ozone origin tracers (see below).

3 Model validation

In the following, previous evalution work concerning the 10 hPa version of the ECHAM/CHEM family is summarised in order to emphasise strengths and weaknesses of stratospheric dynamics and chemistry:

Polar vortex dynamics: Grewe et al. (1998) showed in a detailed analysis of ECHAM3/CHEM that the model is able to simulate a variety of dynamical situations even with a lower vertical and horizontal resolution than now used with E39/C. Vortex erosion caused e.g. by blocking of the flow by the Aleutian high pressure system, vortex splitting, deformation and displacement frequently occurs in the Northern Hemisphere (NH) winter period.

Wind and temperature anomalies: Hein et al. (2001) showed that the zonal mean zonal wind is reasonably well simulated with a clear separation of the subtropical and stratospheric polar night jet, clear inter-hemispheric differences and a reversal of the zonal winds during summer. However, the tilts of the stratospheric jets show differences to the observations and the easterlies during summer do not penetrate deep enough into the lower stratosphere. Both effects are probably linked to the upper boundary since gravity wave effects (dissipation) in the upper stratosphere and mesosphere are not included, i.e. parameterised, like in middle atmosphere models. The evolution of the wind field during the course of the year has been compared to NCEP data by Hein et al. (2001). The extremely low variability during summer is well captured, as well as the extremely high variability during the end of the northern spring (March–May). However, the variability is reduced in E39/C during early winter (December–January). In any case, the variability is large and produces minor warmings with a frequency of about 2 in warm winters. Indeed, major warmings, following the definition of a zonal wind reduction to less than 10 m/s at 30 hPa and 60° N and a reversal of the temperature gradient between 60° N and the Pole at 30 hPa occur 5 times within 20 years of simulation. The increased vertical resolution at tropopause levels clearly increased the model's ability to reproduce the observed inter-annual and intra-seasonal variability (Hein et al., 2001).

Impact of tropospheric variability on stratospheric dynamics: A prominent tropospheric variability pattern is the North-Atlantic Oscillation (NAO). Schnadt and Dameris (2003) showed that the stratospheric response of E39/C to NAO is well simulated in comparison to observations, with a strengthening (roughly 8 m/s increase in zonal wind speed) and cooling (roughly 8 K) of the polar vortex during positive NAO phase compared to negative NAO phase. The heat flux changes are comparable in E39/C and ERA data with respect to pattern and absolute numbers. This clearly indicates that the interaction between tropospheric wave forcing and stratospheric dynamics is reasonably well simulated, which is important to the variability of stratospheric ozone (see below). No indication is found that the upper boundary principally limits these results.

Subsidence: The cooling of the polar vortex results in a descent within the vortex. This has been estimated from various observations to be in the range of 0.05 to 0.08 cm/s at 50 hPa (e.g. see discussion in Grewe et al., 1998, and references therein). E39/C simulates values between 0.08 and 0.1 cm/s, implying that air masses located at 50 hPa at the end of the winter subside from altitudes around 20 to 10 hPa at the beginning of the winter, which still is within the model domain. However, the subsidence is not uniform and the core vortex region has a smaller subsidence. This leads to deficiencies in the evolution of long-lived species, e.g., methane and total chlorine, in the inner polar vortex, especially at the end of winter periods.

Residual streamfunction: Austin et al. (2003) compared a number of climate-chemistry models with respect to the residual streamfunction at 50 hPa. They clearly pointed out that all models (including E39/C) produce the same qualitative pattern as the observations for solstice periods. In northern winter, the results of E39/C agree well with observations between 0 and 45° N. But farther north, the streamfunction gradients decrease significantly, implying less downward transport. The MAECHAM4/CHEM results agree better with observations in high northern latitudes. This could be a result of the higher boundary, though other middle atmosphere models, compared by Austin et al. (2003), are more similar to the values of the E39/C model. Indication that the upper boundary condition is a limiting factor to the simulated stream function are thus weak. This is consistent with

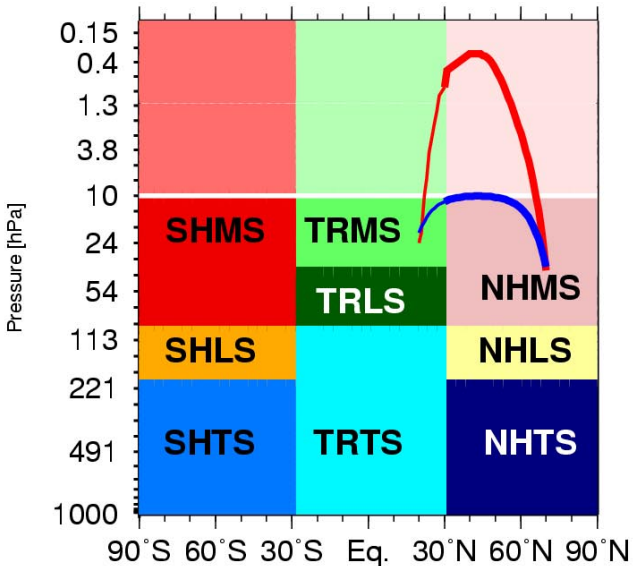

Fig. 1. Nine areas of ozone origin. The colour index is also used for Figs. 5, 8 and 10. See also Table 1. The light shaded areas above 10 hPa (white line) mark the extended regions for MAECHAM4/CHEM. The blue and red lines sketch transport pathways in the two models (see text).

findings of Schnadt et al. (2002), who found an agreement between the pattern of the simulated meridional stream function and derived from observational data between 100 hPa and 10 hPa.

Tropical ascent: An important part of the BDC is the upwelling in the tropics. The 100 hPa upward mass flux for DJF and JJA is simulated to be 19.4 and 16.4×10^9 kg/s, respectively, which is higher than derived from observations (11.4 and 5.6×10^9 kg/s; UKMO; Rosenlof and Holton, 1993) and simulated with middle atmosphere models (13–16.2 and 7.9–9.8×10^9 kg/s, CCM2, Mote et al., 1994) (Stenke, 2006).

Extra-tropical subsidence: In agreement with the overestimated tropical lifting the extra-tropical downward mass exchange is slightly overestimated. Based on UKMO and MLS data estimates of 8.1 and 10.3×10^9 kg/s were given for the NH in DJF, whereas the model simulates 14×10^9 kg/s. In summer observational data give 2.6 and 0.5×10^9 kg/s and the model 5.7×10^9 kg/s (Stenke, 2006).

Age of air: From the analysis of the meridional circulation, it becomes clear that the mean age of air is underestimated in E39/C. In fact, a mean age of air of 3 years is calculated in the tropics at 10 hPa, whereas 3.5 years are derived from observations (Land et al., 1999). It is important to note that although the BDC extends to higher altitudes, the mass included in this circulation, however, is decreasing exponentially.

Ozone profiles: In a variety of papers, ozone profiles have been validated (e.g. Hein et al., 2001). The model E39/C is able to maintain large gradients of various species, e.g. ozone, near the tropopause due to the high vertical resolution (Grewe et al., 2001). Numerical diffusion of ozone caused by the transport scheme is insignificant (Grewe et al., 2002).

Impact of dynamical variability: The dynamical variability of the stratosphere during NH winter periods (e.g. warmings, NAO) has clearly an impact on the ozone chemistry and leads to a variability in total ozone columns of 15% (Hein et al., 2001). The ozone destruction during cold episodes ranges between 10% and 50%, covering well the range derived from observations (see Sect. 5). However, these values are masked by ozone intrusions from lower latitudes into the polar vortex, i.e. the vortex boundary is too leaky. In any case the triggering of different heterogeneously induced ozone destruction through dynamical variability is well captured by the model.

Stratospheric barriers to transport: The stratosphere is characterised by several barriers to transport. The subtropical barrier is clearly visible, e.g. in simulated N_2O fields, and also the frequency of quasi-horizontal exchange processes is realistically reproduced in terms of location, seasonal cycle and absolute frequency (Eyring et al., 2003). However, the polar night jet, as a barrier to transport is too leaky. Although a clear gradient is maintained throughout the winter, subtropical airmasses are mixed into the polar vortex as shown in Sect. 5, masking partially the polar chemical ozone loss.

Inter-hemispheric differences: In general, the polar processing of the chlorine species is well simulated in E39/C, e.g. with respect to the formation of reservoir species $ClONO_2$ and HCl. During Northern Hemisphere winter activated ClO_x is first converted into $ClONO_2$ (including the formation of a chlorine nitrate collar, as observed), whereas during Southern Hemisphere winter the denoxification and denitrification inhibits the $ClONO_2$ formation, so that first HCl is formed (Steil et al., 1998, and references therein).

Impact of variations in tropical ascent on ozone: The interaction between dynamics and ozone chemistry in the lower tropical stratosphere, as simulated by E39/C, has been described in Stenke and Grewe (2005). After the eruption of the Mt. Pinatubo, the tropical ascent is increased, which leads to a shift in the ozone profile in the region where ozone is predominntly dynamically controlled. The resulting ozone deline of 4–5% agrees well with observations (Stenke und Grewe, 2005, and references therein).

QBO and solar cycle: Recently, Dameris et al. (2005) and Steinbrecht et al. (2006) presented an evaluation of E39/C (Steinbrecht et al., includes a comparison to MAECHAM4/CHEM) for the transient evolution between 1960 and 1999, with special emphasis on the variability of stratospheric ozone. In principle MAECHAM is able to internally simulate a realistic QBO (Giorgetta et al., 2002), however, then 90 levels are required, which inhibits multi-annual simulations. Therefore, Dameris et al. (2005) and Steinbrecht et al. (2006) applied a relaxation methodology

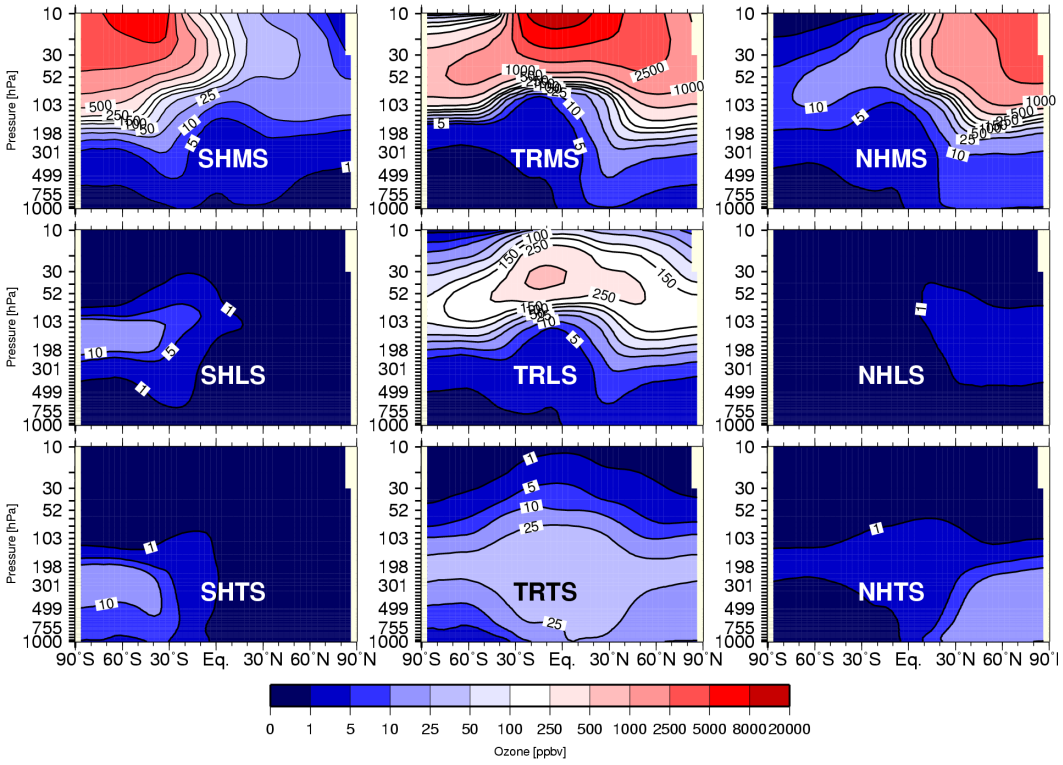

Fig. 2. Contributions of the nine ozone production regions to the total ozone content [ppbv] as simulated with E39/C for January.

("nudging") for equatorial winds to correctly represent QBO effects. Solar cycle effects were represented by varying the solar constant (affecting the heating rates) and extra terrestrial fluxes (affecting the photolysis rates). Both models revealed a good agreement with observational data throughout the period. The solar flux changes are inducing ozone changes, which are in the order of 3% (see Dameris et al., 2005, Fig. 15), in agreement with observations. The propagation of a solar cycle effect to higher latitudes is simulated in both models (Steinbrecht et al., 2006, Fig. 5). Similar results can also be found with respect to QBO effects. Equatorial westerlies are correlated with positive ozone anomalies at tropical latitudes and high latitudes and negative ozone anomalies at mid-latitudes. This pattern can be found in both model versions as well as in observational data. These effects are not included in the present paper, but the comparison shows that the E39/C model is able to react to external and internal forcings in a reasonable way.

Combined tropospheric-stratospheric effects: Another important phenomenon, which is a combined effect of tropospheric and stratospheric dynamics, is the occurrence of so-called ozone miniholes. Poleward moving tropospheric tropical high pressure systems are accompanied by a high tropopause and lead to extremely low total ozone values, especially when they are located below the polar vortex, because they induce a divergent stratospheric flow. Stenke and Grewe (2004) showed that this interplay of tropospheric and stratospheric dynamics on ozone is well simulated by E39/C in comparison to TOMS satellite data, in terms of location and seasonal cycle of occurrence: maximum occurrence is found over Europe, Greenland and North-west America with a peak in January and February.

Clearly, the low upper boundary of the E39 climate model leads to differences in reproducing stratospheric dynamics compared to middle atmosphere models and observational data. The model shows deficiencies in the mean age of air, tropical ascent and polar subsidence. However, none of the findings indicate that this upper boundary is a principle and thorough restriction to the model, which is preventing its use with respect to troposphere and lower stratosphere dynamics. Comparisons to middle atmosphere models did not indicate that among the limitations of all these models, the upper boundary is an outstanding issue. These results indicate that the E39/C model is able to simulate tropospheric and stratospheric ozone with respect to a mean state but also and that is important in the context of the present paper, with respect to variability.

4 Methodology

As a first step, areas of interest for the ozone origin are defined (Table 1). These nine areas (3 latitude bands and 3 height regions) are illustrated in Fig. 1. The areas are

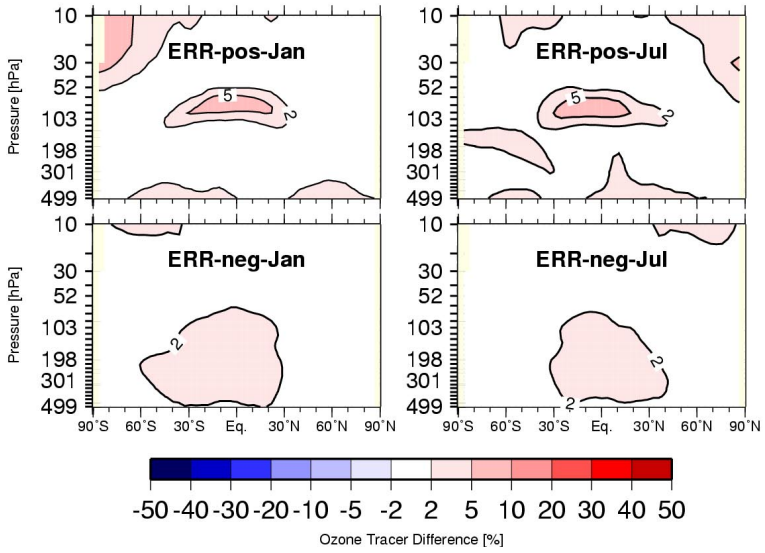

Fig. 3. Calculated errors (top: positive, bottom: negative) associated with the methodology relative to the ozone background (%) for January (left) and July (right).

chosen, so that the main ozone production area (tropical mid-stratosphere) can be separated from extra-tropical regions and that the tropospheric ozone production areas can be separated from the stratosphere. For the MAECHAM4 version the regions SHMS, TRMS and NHMS are extended to 0.01 hPa, indicated by the lighter colours. An accurate separation of the areas according to the location of the tropopause has not been chosen because the ozone production rates in those regions are small and the changes to the total burden of one of the nine tracers are small too. At mid-latitudes, the results show that ozone from those regions have only a minor contribution (see Sect. 5). In the tropics the tropopause show only small variations and remains mainly in one model layer, which is then identical to the definition of the areas in Fig. 1 anyway.

In a second step, an ozone tracer ($^{i}O_3$, with i index for the region) is assigned to each of the nine regions. Each tracer experiences ozone production only in that region, but ozone destruction in the whole model domain, according its concentration:

$$\frac{d\,^{i}O_3}{dt} = {}^{i}O_3{}^{\mathrm{Prod}} - O_3{}^{\mathrm{Loss}}\,\frac{^{i}O_3}{O_3}, \qquad (1)$$

with $^{i}O_3{}^{\mathrm{Prod}}$ and $O_3{}^{\mathrm{Loss}}$ the ozone production and loss in $m^3/m^3/s$, derived from the modelled ozone O_3 in m^3/m^3, with the restriction that $^{i}O_3{}^{\mathrm{Prod}}$ equals to zero for each grid point outside the region i. It can easily be shown that $\sum_i {}^{i}O_3 = O_3$ for all time steps and grid points, if this equation holds for the initial conditions and that for all other initial conditions $\sum_i {}^{i}O_3$ converges exponentially, proofing the convergence of the methodology. At any grid point, ozone originates therefore by $\frac{^{i}O_3}{O_3}\times 100\%$ from region i.

Since the different gradients of the ozone tracer may produce a different numerical diffusion, the sum of all tracer may not add up to the simulated ozone. The ozone origin tracers are therefore linearly scaled at every timestep to overcome this deficiency. This potential error can be quantified since

$$\frac{d}{dt}\sum_i {}^{i}O_3 = \frac{d}{dt}(O_3 + \epsilon^{+} - \epsilon^{-})$$
$$= (O_3{}^{\mathrm{Prod}} + \Delta^{+}) - \left(\frac{O_3{}^{\mathrm{Loss}} + \Delta^{-}}{O_3}\right)(O_3 + \epsilon^{+} - \epsilon^{-}), \qquad (2)$$

where O_3 is the simulated ozone, Δ^{+} and Δ^{-} the positive and negative errors produced at every timestep in $m^3/m^3/s$, $\epsilon^{+} \geq 0$ and $\epsilon^{-} \geq 0$ the accumulated errors in m^3/m^3. The differential equation can be splitted up into two equations:

$$\frac{d}{dt}\epsilon^{+} = \Delta^{+} - \left(\frac{O_3{}^{\mathrm{Loss}} + \Delta^{-}}{O_3}\right)\epsilon^{+} \qquad (3)$$

$$\frac{d}{dt}\epsilon^{-} = \Delta^{-} - \left(\frac{O_3{}^{\mathrm{Loss}} + \Delta^{-}}{O_3}\right)\epsilon^{-} \qquad (4)$$

This analysis technique has been implemented into the climate-chemistry model E39/C (Hein et al., 2001) and applied to a model simulation, which is identical to Grewe et al. (2001) and to the MAECHAM4/CHEM model. Figure 2 shows the results for E39/C for January. For the region TRMS (mid of the top row) it can clearly be seen that ozone is produced in that area and how it is destroyed along its way through the atmosphere. Only roughly 1 to 5 ppbv out of 6–9 ppmv, i.e. less than 0.1%, is reaching the ground in the Northern Hemisphere. It does not imply that these ozone molecules are actually produced in January, indeed

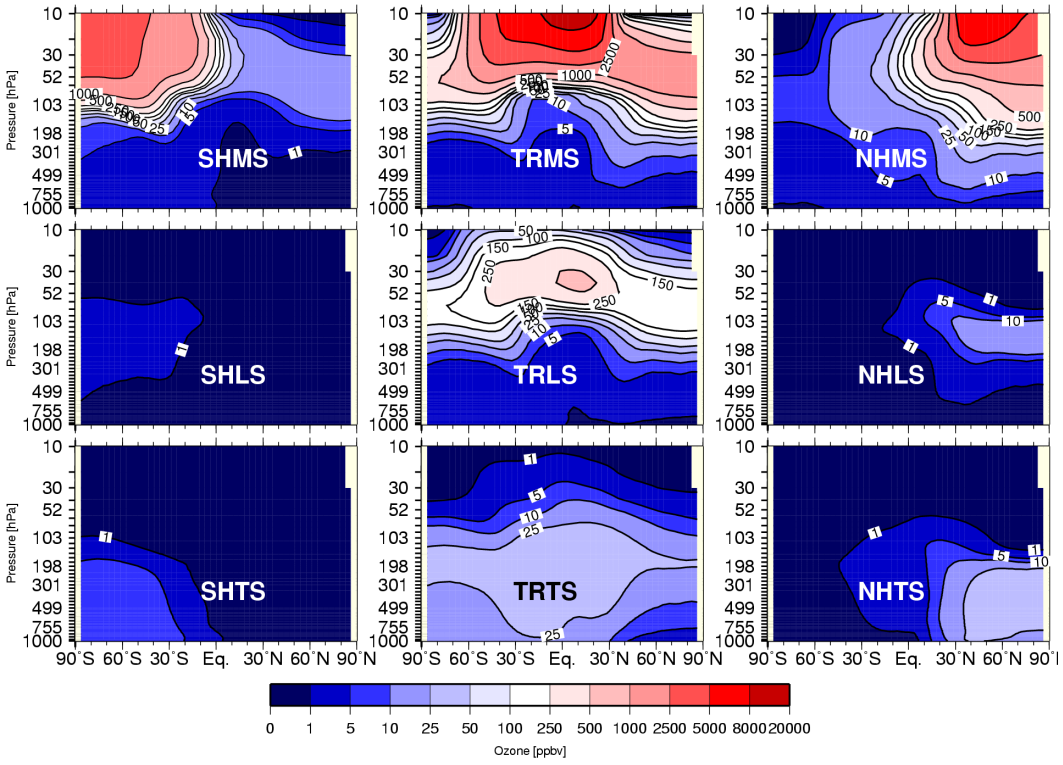

Fig. 4. Contributions of the nine ozone production regions to the total ozone content (ppbv) for July calculated with E39/C.

more likely during earlier months, i.e. that they are a reminiscence from e.g. summertime ozone production, which is then transported slowly into the troposphere.

The errors associated with this methodology (ϵ^+ and ϵ^-) are shown in Fig. 3 for January and July. In most regions, the accumulated positive and negative errors are less than 2%. Only around the tropical tropopause and at high latitudes the error exceeds 5%.

5 Origin of stratospheric ozone

Figures 2 and 4 show the evolution of the nine diagnostic ozone tracers in E39/C for January and July, respectively. The first impression is that each ozone tracer has its peak value in the region of its origin, which directly follows from Eq. (1). In the tropics at 10 to 30 hPa, ozone is mainly chemically controlled with a lifetime between 10 (at 10 hPa) and 100 (at 30 hPa) days, which implies that local ozone production plays the major role. However, at mid and higher latitudes the picture is reversed. Between 30° N and 60° N in January and between 30° S and 45° S in July, i.e. in the winter hemispheres, the tropical ozone (TRMS) dominates over local ozone production. This can be made clearer by colour coding the regions of origin and re-arranging them in the order of their importance (Fig. 5): first (second, third) column shows the main (second, third) contributing region, indicated by the colour code (Fig. 1). Isolines indicate how much the region contributes to the ozone amount (in percentage). For example, in January at 20 hPa and 40° N, i.e. in the region NHMS, the main contributor to the ozone concentration is the ozone produced in the tropics (light green, TRMS) with 70%, followed by ozone produced in the NMHS region (pink) by around 30%. Other contributors are almost unimportant with values less than ≈2% (e.g., dark green, TRLS). However, in the summer hemispheres ozone chemistry is fast enough, i.e. the life-time is small enough, that the local ozone production dominates over transport from the tropics. This means that the seasonal cycle is an important factor for regenerating stratospheric ozone at mid and high latitudes. Throughout the year, regional ozone production dominates over transport from tropical regions for ozone north of 65° N and above 40 hPa, which clearly contradicts to the idea of the tropical ozone production being the main source for stratospheric ozone.

In the following, a comparison of these findings to results from the MAECHAM4/CHEM model is provided in order to better understand the impact of the E39/C's upper boundary condition. Figure 6 shows the nine ozone tracers for January simulated with MAECHAM4/CHEM. Clearly, the structures below 10 hPa (indicated by a horizontal line) are similar to E39/C (Fig. 2). The cross tropical transport is somehow larger in the middle atmosphere model between 50 hPa and 100 hPa, e.g. values between 100 and

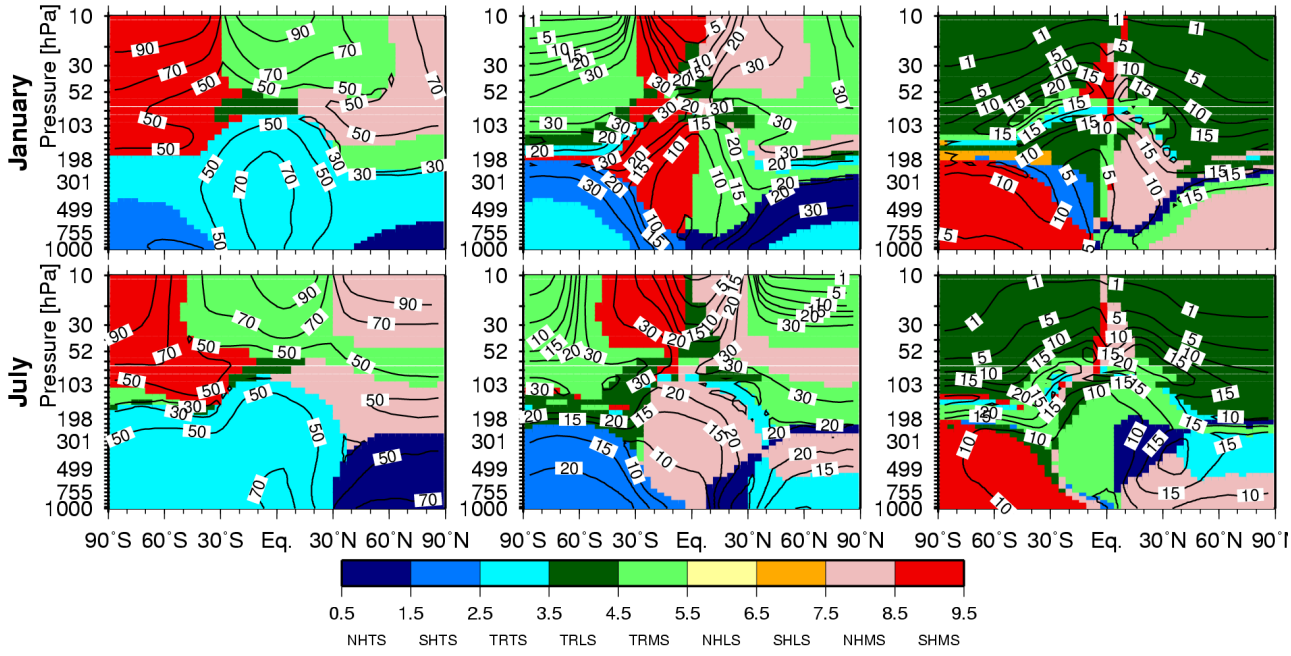

Fig. 5. Decomposition of the zonal mean ozone content. Main contribution (first column), second (second column) and third (third column) important contribution to the ozone content for January (top) and July (bottom). The colour code (see Fig. 1) indicates the origin, the isolines its mixing ratio (ppbv). Isolines are only meaningful within one colour area.

150 ppbv can be found at northern mid-latitudes, whereas E39/C shows values around 25 pbbv. Figure 7 shows the differences in the relative contributions from the individual tracers between MAECHAM4/CHEM and E39/C, i.e. $\left(\frac{^{i}O_3}{O_3}\times 100\%\right)^{E39/C} - \left(\frac{^{i}O_3}{O_3}\times 100\%\right)^{\mathrm{MAECHAM4/CHEM}}$, for the three main stratospheric tracers (SHMS, TRMS, and NHMS). The differences are for most regions in the order of 5%. However, at some regions the differences reach 20% to 30%. E39/C simulates more TRMS-tracer mass at winter-time mid-latitudes, around 20–30% in January and 10–20% in July (mid row). This is caused by the shorter transport times of ozone from the tropics to the mid-latitudes in E39/C, which is indicated in Fig. 1 (thick blue lines) compared to MAECHAM4/CHEM (longer thick red line). Since the transport time to that region is longer in MAECHAM4/CHEM this ozone tracer is more efficiently destroyed. This means that the contribution of low latitude ozone to the ozone concentration at higher latitudes is even overestimated in E39/C. This also implies that in E39/C the contribution of other ozone tracers has to be lower. That is the local ozone production, which has a lower contribution to ozone at that region (see green arrows). The second difference occurs at the summer pole, where E39/C simulates more ozone from local ozone production and less from mid-latitudes (red arrows). This is consistent with higher cross equator transport in MAECHAM4/CHEM, discussed above. One hardly can conclude, which of both models represents this cross equator exchange of ozone at pressure levels between 10 and 50 hPa more realistic, whereas the MAECHAM4/CHEM certainly better represents the BDC at altitudes above 25 hPa. Except for these two differences, which have to be taken into account in the further analysis, the results indicate that E39/C leads to similar findings as the middle atmosphere model MAECHAM4/CHEM. Thus both models indicate that stratospheric ozone at higher latitudes is dominated by ozone production in that region rather than transport from tropical regions.

As concluded above, the seasonal cycle plays an important role for the origin of ozone. Figure 8a illustrates the seasonal cycle for the main contributor to ozone at 50 hPa, 60° N, i.e. at the edge of the region, where ozone predominately originates from the NHMS region throughout the year. There, ozone originates mainly from the tropical mid-stratosphere between February and July, with peak values of 2.2 to 2.7 ppmv in March and April, well-known as the spring-time mid-latitude ozone maximum. These high values are then reduced by roughly 50% to 60% until October, reaching then values of about 1 to 1.2 ppmv. Ozone produced in the extra-tropics decreases from higher values in winter (2.3 to 2.5 ppmv) down to about 1.4 ppmv in June, indicating that local ozone production dominates over transport from the tropics between August and February. In the beginning of the winter season (November) the mid-latitude ozone concentration originates by approximately 1/3 from the tropics and 2/3 from local ozone production during summer and autumn. Contributions from other regions are almost unimportant.

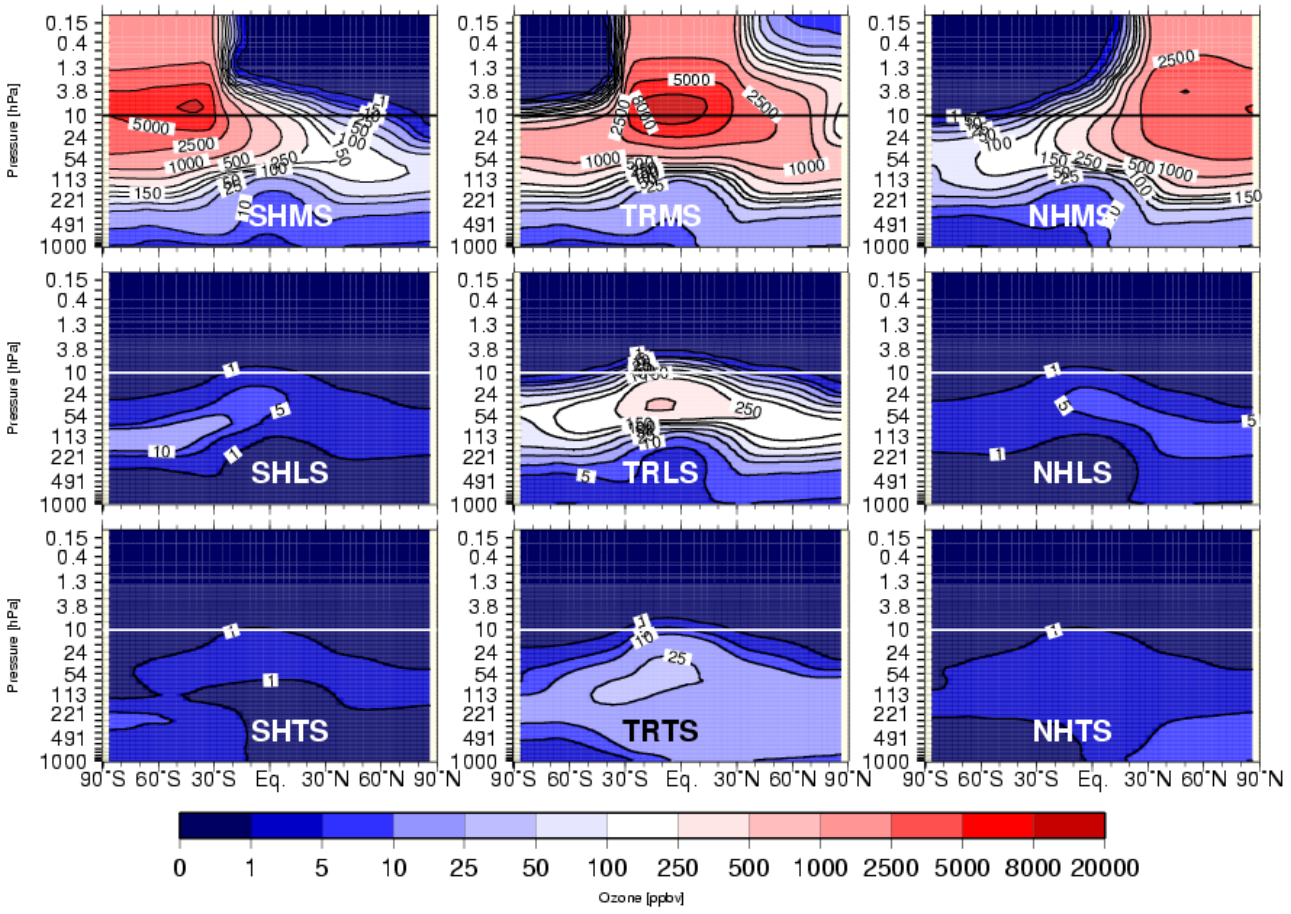

Fig. 6. Contributions of the nine ozone production regions to the total ozone content [ppbv] for the middle atmosphere model MAECHAM4/CHEM for January.

At polar latitudes (90° N; Fig. 8b), the picture is somewhat different. In the beginning of the winter, ozone originates mainly from local (NHMS) ozone production with around 3 ppmv, which is then chemically destroyed, reaching values in February and March of about 1.5 to 2.7 ppmv, showing a loss of about 0.3 to 1.5 ppmv. The large inter-annual variability is consistent with observational data and covers the observed range (Müller et al., 1996; Rex et al., 2004). It shows the ability of the model E39/C to simulate the variability of the stratospheric chemical and dynamical processes. Ozone originating from the tropics has an increasing importance at high latitudes from January (≈1 ppmv) to April (up to 3 ppmv). To better illustrate the chemical regime and to better separate dynamical from chemical processes, Fig. 8c shows the seasonal cycle at 90° N equivalent latitude, i.e. for the centre of the polar vortex, instead of the North Pole like in Fig. 8b. In the centre of the vortex, the increase of tropical ozone is delayed compared to the North Pole, and the high TRMS ozone values of up to 3 ppmv are only reached at the end of the winter, when the vortex breaks up and mixing of vortex and non-vortex air is increased. Wintertime ozone supply from the tropics is increasing very differently from year to year between roughly 1 and 2 ppmv, as a consequence of the inter-annual variability in simulated dynamics and transport. It also partially masks the chemically ozone loss of 0.3 to 1.5 ppmv in the model, so that the ozone depletion is correctly simulated, but the total ozone decline ends up to be underestimated. Ozone produced in the TRMS region declines then quite rapidly from March onwards mirroring the decrease of the ozone life-time. This is in accordance with previous findings by Stenke and Grewe (2004), who showed that the life-time of a spring-time chemically induced ozone perturbation (in this case caused by ozone mini-holes) vanishes totally at the end of summer. Fioletov and Shepherd (2003) found similar values for the photochemical relaxation times for Northern Hemisphere mid-latitude total ozone. They showed that ozone anomalies in late spring are correlated with summertime ozone. The correlation still holds until beginning of autumn, with low values however. Deviations in March of ±30 DU decrease to ±20 DU in June and ±5 DU in September, based on TOMS and SBUV data from 1979 to 2000. This would suggest an e-folding time of

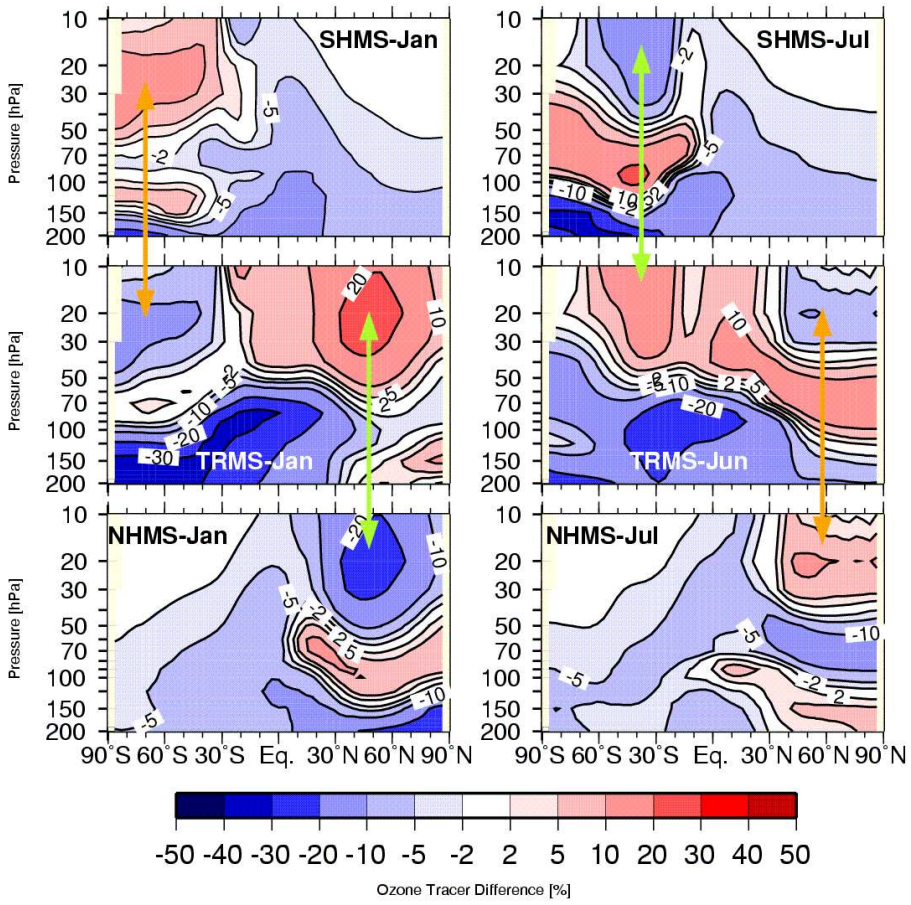

Fig. 7. Differences (%) in the contribution of the ozone tracers SHMS (top), TRMS (mid), and NHMS (bottom) between the models E39/C and MAECHAM4/CHEM for the region 200 hPa to 10 hPa for January (left) and July (right).

around 3 months, similar to the simulated values, though the approach differs.

At lower stratosphere tropical altitudes (100–60 hPa, Fig. 8d) ozone chemistry is slow and ozone is dominated by transport rather than by chemistry (Brasseur and Solomon, 1986). Since the simulated meridional circulation has also a seasonal cycle, which peaks in February (Reithmeier, 2001), the impact of tropospheric ozone (TRTS) peaks with some delay in April. The contribution from NHMS region has a quite strong seasonal cycle, peaking in June, when it contributes with similar amount to local ozone like the local ozone production. This reflects the BDC, which transports ozone towards the winter poles, i.e. ozone produced at mid and high northern latitudes during March to June is transported to lower latitudes. In total, up to 0.5 ppmv (Figs. 2 and 4) can be produced locally and transported to higher latitudes via streamer events (Eyring et al., 2003).

In the extra-tropical tropopause region, ozone, which is produced in the TRLS region and transported to higher latitudes via streamers foms an important contribution to the ozone concentration of at least 20% (Fig. 5). In the Southern Hemisphere, this is the second important contributor with roughly 30%.

6 Stratosphere-troposphere exchange

Recently, Stohl et al. (2003) gave an overview on the processes important to stratosphere to troposphere exchange. It is known for long that the large-scale stratosphere-troposphere exchange of air mass occurs predominantly in conjunction with the subtropical jet stream and the polar jet stream (Danielsen, 1968; Holton et al., 1995). Figure 9a shows the simulated mass exchange (net flux) calculated with the method of Wei (1987), using pressure as vertical coordinate as suggested by Grewe and Dameris (1996). The WMO criteria (lapse rate) is taken for identifying the tropopause. The values are in good agreement with those derived with the same methodology based on meteorological data from the European Centre of Medium-Range Weather Forecasts (Grewe and Dameris, 1996). However, the accuracy of the Wei formula has been under debate, the net flux is a residuum and depends additionally on the coordinate

system (Grewe and Dameris, 1996; Wirth and Egger, 1999; Stohl et al., 2003). Reithmeier (2001) compared the net STE derived from the Wei formula with a flux estimate derived from a Lagrangian approach, taking into account exchange events lasting at least 12 h. The differences between these methods were in the range of 10 to 20% for most areas for DJF. Sprenger and Wernli (2003) presented a climatology of zonally integrated net cross tropopause mass fluxes, which are significantly lower than those those obtained here. However, the approach differs: Sprenger and Wernli (2003) showed deep intrusions, with a residence time of at least 96 h, whereas here 12 hourly data are applied, this may well explain half of the discrepancy (Wernli and Bourqui, 2002).

The simulated stratosphere-troposphere exchange occurs clearly in conjunction with the sub-tropical jet-stream, peaking on either hemisphere in winter and spring. Since the location of the jet stream moves to higher latitudes in summer in either hemisphere, the air mass exchange peaks also at higher latitudes.

The respective ozone fluxes are shown in Fig. 9b. A total annual mean ozone flux into the troposphere of 420 TgO_3/a (range: 346–474) is simulated, which is on the lower side of estimates derived from observations of 450 (range: 200–870) TgO_3/a (Murphy and Fahey, 1994), 510 (range: 450–590) TgO_3/a (Gettleman et al., 1997) and 500 (range: 479–523) TgO_3/a (Olsen et al., 2004). However, compared with other modelling studies, ranging from 391 to 1440 TgO_3/a (IPCC, 2001), and taking into account the estimate uncertainties and ranges, the simulated ozone influx of 420 Tg/a seems to be reasonable, although one has to take into account that the uncertainty associated with the methodology are still to be quantified (see above). The overall pattern of the ozone flux is similar to the mass fluxes. However, the seasonal cycle of ozone in the lower stratosphere has an impact on the absolute values, e.g. leading to higher ozone fluxes at around 30° N during spring (MAM) compared to winter (DJF).

Figure 10 shows where the exchanged ozone originates from for the Northern (a) and Southern (b) Hemisphere. In the Northern Hemisphere the annual influx is dominated by ozone produced in the NHMS region. It shows a quite pronounced seasonal cycle, with lowest values in winter, because of the low ozone production rates and the stronger impact from the tropics, since the ozone life time is increased, leading to farer-reaching dispersion of tropical ozone. For the same reason ozone from the TRMS region has a minimum in summer-time. Almost the same amount of ozone mixed into the troposphere originates from the tropical mid-stratosphere (TRMS) and from the tropical troposphere (TRTS). Latter represents a re-circulation through the lowermost stratosphere.

Exactly this mechanism leads to the most important contribution to the ozone influx in the Southern Hemisphere (Fig. 10b). As in the Northern Hemisphere, the seasonal cycle of the ozone mixed into the troposphere originating from

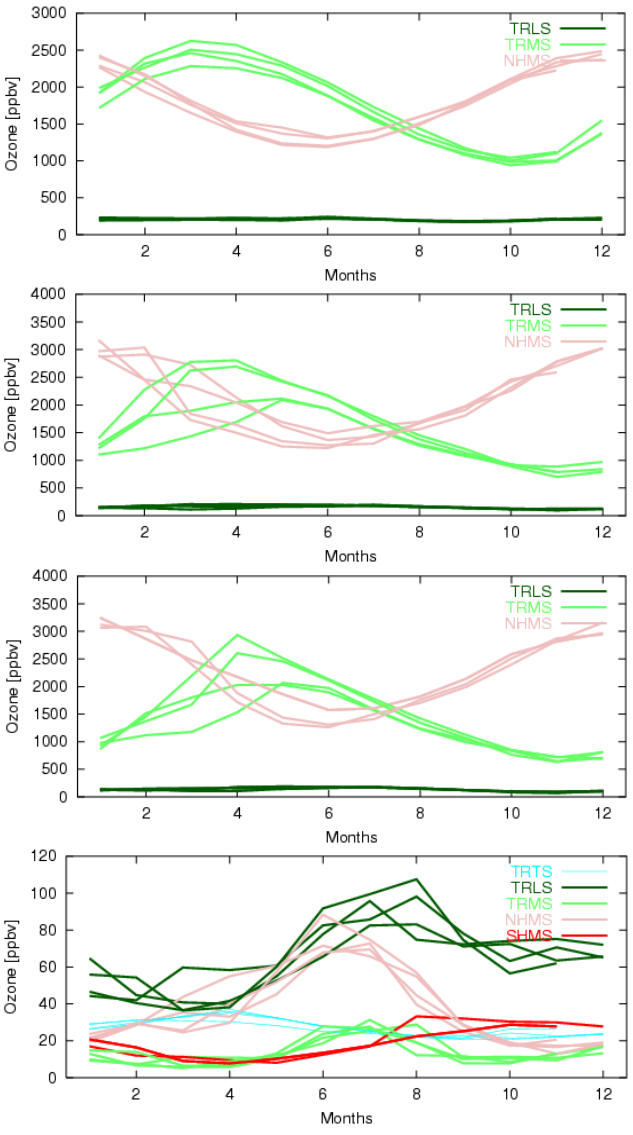

Fig. 8. Seasonal cycle of the ozone mixing ratios (ppbv) from various origin at 60° N and 50 hPa (first row), 90° N and 50 hPa (second row), 90° N equivalent latitude and 50 hPa (third row), and the Equator at 76 hPa (bottom row). Four consecutive cycles are presented.

SHMS is reversed compared to those from the tropics (TRTS, TRLS, and TRMS) again for the same reasons.

7 Origin of tropospheric ozone

In the troposphere the origin of ozone at low altitudes is mainly dominated by local ozone production (Fig. 5), except for the ozone in the southern extra-tropics during winter, where the transport of tropical ozone is over-compensating the local ozone production. In the tropics, ozone chemistry is fast enough throughout the year so that the local

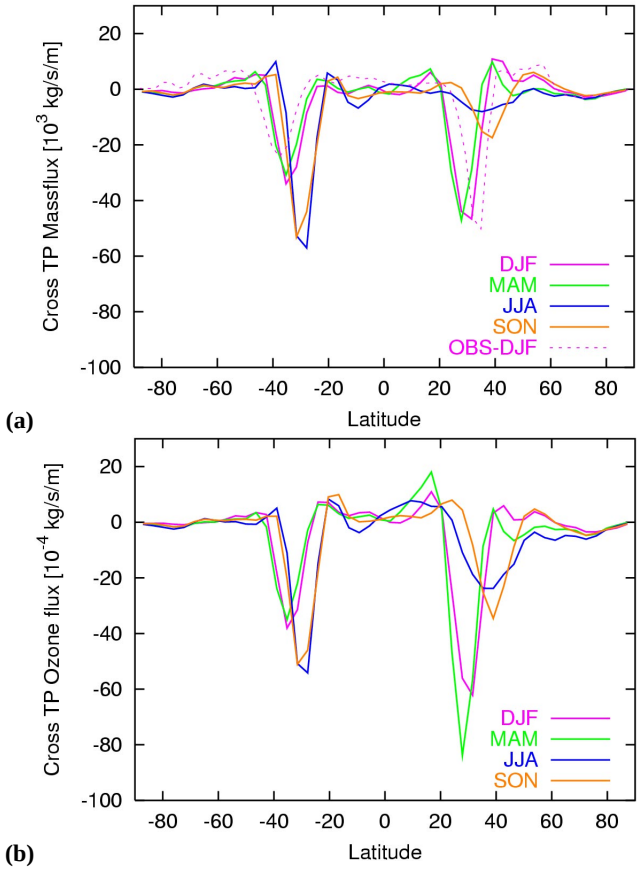

Fig. 9. Zonal mean air mass **(a)** and ozone **(b)** exchange (kg/m/s) through the thermal tropopause for the 4 seasons. The dashed line indicates DJF mass exchange derived from ERA-15 data, taken from Grewe and Dameris (1996).

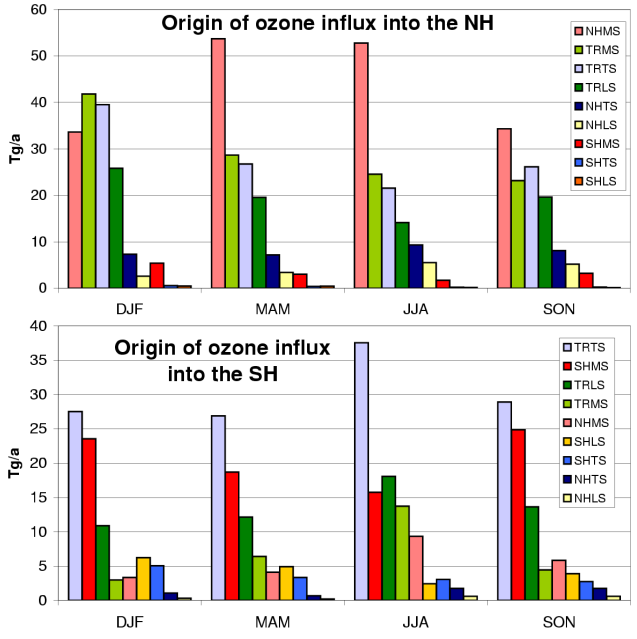

Fig. 10. Ozone influx from the stratosphere into the troposphere (Tg/a) for the Northern (top) and the Southern Hemisphere (bottom) splitted up into the regions of ozone origin. The regions are in the order of their importance regarding the total annual influx.

ozone production is always more important than ozone transport from higher latitudes or from the stratosphere. Interestingly, largest contributions to the local ozone concentration in the upper troposphere of both hemispheres are coming from tropical ozone (TRTS) throughout the whole year, except for northern summer (Fig. 5, left), where ozone from lower altitudes dominates. At lower altitudes, tropical ozone is even second important in the extra-tropics, probably it is mixed down from the upper troposphere to the lower troposphere, since low level transport would be combined with a much shorter ozone lifetime.

Ozone, which is produced in the stratosphere (i.e. not in NHTS, TRTS, and SHTS), has an important contribution to ozone in the troposphere, in the order of 30% in the winter hemisphere and 20% in the summer hemisphere (Fig. 11). However, the contribution is systematically higher (around 5%) in the Northern Hemisphere than in the Southern Hemisphere, for the respective season. This largely is a consequence of the higher ozone exchange rates in the Northern Hemisphere compared to the Southern Hemisphere (Fig. 9). Ozone in the tropical troposphere clearly has a lower contribution from ozone produced in the stratosphere because of faster chemistry and, perhaps even more important, less significant influx from the stratosphere. The areas with stronger ozone influx (Fig. 10) clearly show an enhanced stratospheric contribution. The origin of ozone produced in the stratosphere and found in the troposphere can be identified in Fig. 5. In the tropics, ozone mainly originates from SHMS and TRMS in January and from NHMS in July. In the extratropics, the most important regions are SHMS for the Southern Hemisphere and NHMS for the Northern Hemisphere, but not the region TRMS. This again indicates that extratropical ozone production dominates over tranport of ozone from the tropics.

Since non-methane-hydrocarbon (NMHC) chemistry is not included in the model, the ozone lifetime is overestimated by the model, which likely leads to too high values of stratospheric ozone found in the troposphere, especially close to the Earth's surface. Lamarque et al. (2005) applied the MOZART-2 model, which includes NMHC chemistry. They found a contribution of stratospheric ozone to the surface ozone concentration of up to 10% in the Northern Hemisphere and up to 20% in the Southern Hemisphere, for 1990 conditions. Whereas in this study the contributions are as much as 20% in the Southern Hemisphere and around 25% in the Northern Hemisphere, which partly (besides differences in emission datasets) can be explained by the enlarged ozone lifetime in the E39/C model near the surface. On the other hand, Kentarchos and Roelofs (2003) present simulated

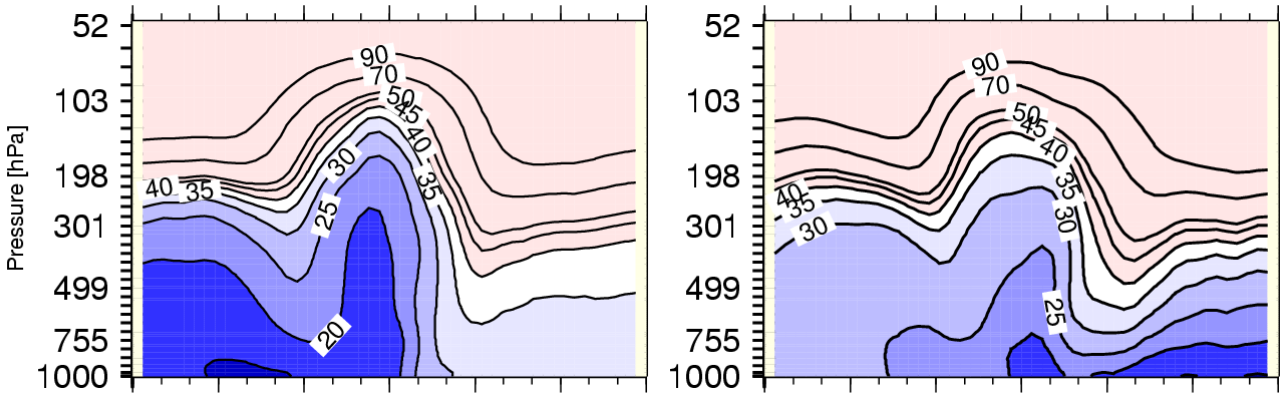

Fig. 11. Contribution (%) of stratospheric ozone to the total ozone for January (left) and July (right) calculated with E39/C.

stratospheric ozone in the troposphere for April with values around 5 and 10 to 15 ppbv in the Southern and Northern Hemisphere, respectively, which agrees well with E39/C values (not shown). However, Kentarchos and Roelofs (2003) estimate the stratospheric ozone flux by 641 Tg/a, which could indicate that a smaller STE flux of ozone and a longer ozone lifetime in E39/C lead the same near surface ozone concentrations as in their model, given that the STE flux estimate can be compared.

8 Discussion

In order to determine the origin of ozone, climate-chemistry models were applied and extended by to a simple diagnostics. That implies that the quality of the results are largely determined by the quality of the model and the diagnostics.

First, the diagnostics, which marks ozone molecules according to their region of production (see Sect. 4) has a very low potential error, since ozone transport is linear and chemistry is not affected. The only potential error source is numerical diffusion, caused by the transport scheme in situations, where gradients are large. However, the relatively high vertical resolution at tropopause regions of about 700 m, is sufficient to correctly maintain gradients especially at tropopause altitudes, where the ozone gradient maximises (Grewe et al., 2002). These errors are quantified (Eqs. 3 and 4) to be in the order of 2% (Fig. 3).

Second, the model's ability to simulate atmospheric transport and chemistry, and its interactions has to be good enough to come to reliable conclusions. From a detailed evaluation of the E39/C model (Sect. 3), the main drawbacks were identified as (1) too fast tropical ascent, (2) too fast transport from the equator to the winter pole, related to the low upper boundary, since the upper branch of the BDC is bundled in the upper most levels and (3) too leaky polar vortex boundaries. The latter has been analysed by using the ozone origin analysis, demonstrating the ability of the applied methodology to identify model deficits. On the other hand, the interaction of chemistry and transport in the extra-tropical stratosphere seems to be reasonable, since the turn-around time of a stratospheric ozone perturbation is realistically simulated (see Sect. 5).

To better identify the model dependency of the results for the stratosphere, a middle atmosphere climate-chemistry model has been applied. In many regions the results do not differ significantly from E39/C. However, E39/C simulates a too fast poleward transport, which leads to an overestimate of the impact of tropical ozone on higher latitudes. This implies that the one finding of this paper, namely that less than 50% of the ozone found at higher stratospheric latitudes is actually produced in the tropics, is even an upper boundary, and the MAECHAM4/CHEM results point at 30%, because longer transport time-scales decrease that contribution.

The evaluation of the ozone flux from the stratosphere to the troposphere is a challenge. In Sect. 6 it has been shown that the exchange of ozone into the troposphere seems to be reasonably simulated, at least within the ranges of uncertainty. However, there is still the need to assess the uncertainties of the methodologies and to derive a consistent way of comparing values derived from observations with model data. How much the ozone, produced in the stratosphere, contributes to the tropospheric ozone budget is a combination of tropospheric mixing and chemical ozone lifetime. At the surface, observational data suggests that the stratospheric contribution amounts to 14% at Areskutan, Sweden, with peak values up to 50% (Bazhanov and Rodhe, 1997). Model studies suggest a maximum contribution to near surface ozone ranging from 5% (Follows and Austin, 1992) to 10% (Southern Hemisphere) and 20% (Northern Hemisphere) (Lamarque et al., 2005). This is less than found in this study (15%–30%). It may partially be explained by missing NMHC chemistry, which would increase tropospheric HO_2 concentration and therefore reduce the ozone lifetime (Roelofs and Lelieveld, 2000). Another model study suggests that the stratospheric ozone may contribute to the tropospheric ozone column by 30% to 70% (de Laat et al., 2005),

which seems to be more in agreement with our findings and indicates that the ozone lifetime may be overestimated especially at lower altitudes. Marcy et al. (2004) gave estimates of the stratospheric contribution to upper tropospheric ozone based on very precise HCl measurements, which correlate to stratospheric ozone. They concluded that 20% to 80% of the upper tropospheric ozone (10 to around 15 km at 31° N–33° N) originate from the stratosphere. Taking into account the large variability in the region of the subtropical jet, where the measurements were performed, those values are well covered by the model. However, a more climatological basis would be needed to sufficiently evaluate the model.

Comparing Fig. 2 and Fig. 6 gives an indication on the different importance of stratospheric ozone in the troposphere in the model MAECHAM4/CHEM and E39/C. In mid and high latitudes of both hemispheres the middle atmosphere model simulates a contribution of roughly 70% at 5 km, which is far more than in E39/C (SH:≈20%; NH:≈30%). Clearly the stratosphere to troposphere exchange of ozone is significantly larger in the middle atmosphere model compared to E39/C. An enhanced stratosphere to troposphere air mass exchange has already been reported earlier for MAECHAM4 (Meloen et al., 2003; Cristofanelli et al., 2003). This also shows the advantage of a higher resolution at tropopause levels, leading to a better representation of the stratosphere to troposphere exchange, which agrees with earlier findings (Timmreck et al., 1999; Grewe et al., 2002).

9 Conclusions

In this study, the origin of ozone has been analysed, by applying the climate-chemistry models E39/C and MAECHAM4/CHEM and including regionally marked ozone tracers. This methodology allows to attribute the ozone concentration at a given point of the atmosphere to some regions, where these ozone molecules were generated, i.e. where its origin is located.

E39/C has the advantage to resolve both, troposphere and stratosphere dynamics and chemistry. It is designed with an emphasis to reasonable transport in the transition zone, e.g. by an increased vertical resolution at tropopause levels compared to the standard ECHAM version. However, its upper level is centered at 10 hPa, which bundles the upper stratosphere circulation in the uppermost levels. To better understand the impact of this drawback, the middle atmosphere chemistry-climate model MAECHAM4/CHEM has been applied. Basically, it confirmed the results obtained by E39/C, e.g. concerning the evolution and pattern of ozone origin tracers in the stratosphere.

Although highest local ozone production rates can be found in the tropics at around 30 km, this region does not dominate the extra-tropical stratospheric ozone distribution. At higher latitudes the local ozone production rates are considerably smaller, but the destruction rates are also smaller and, especially in summer-time, photochemical ozone production is the dominant ozone source. This leads to a contribution of tropical ozone to the extra-tropical ozone content of less than 50%, which is an upper boundary, since this value is even less in the middle atmosphere model (30%).

During winter and spring ozone within the polar vortex, predominantly originates from NHMS ozone production during the preceding summer and autumn. Ozone mixing ratios of about 3 ppmv are depleted in E39/C during winter and spring to values as low as 1.5 ppmv, i.e. a maximum ozone destruction of up to 50% is found. Ozone supply from the tropics (TRMS) is increased during that time, partially masking the chemical ozone depletion, which may be a result of the location of the uppermost model layer at 30 km. Here it is noteworthy to point out that this model deficiency could only be identified through the applied methodology.

The Northern Hemisphere is characterised by large dynamical variability during winter and spring. This produces a large variability in PSC formation and polar ozone destruction and results in large inter-annual variations of ozone, which is also reflected in the exchange of stratospheric ozone into the troposphere. The total influx of ozone into the troposphere of around 420 TgO_3 per year has been found to agree fairly well with estimates derived from observations and other model studies. However, it has to be noted that the methodologies to estimate this quantity differ and that the used methodology is less accurate than e.g. Lagrangian approaches. In the Northern Hemisphere the region NHMS dominates this influx with contributions from TRMS (tropical mid-stratosphere) and TRTS (tropical troposphere). The latter means a recycling of tropospheric ozone through the lower-most stratosphere. This recycling process is found to be the dominant source for ozone mixed into the troposphere in the Southern Hemisphere.

Surface ozone is found to be produced locally by around 75% and 85% in winter and summer, respectively, which may be underestimated due to the lack of NMHC chemistry in E39/C. However, other model studies (Lamarque et al., 2005) suggest that the differences are not large and may be in the order of 10%.

All these findings indicate that the view of the tropical region as the global source for stratosperic ozone, is highly questionable. The results indicate that the tropics indeed contribute to extra-tropical stratospheric ozone, but by far more important is the extra-tropical ozone production by itself.

In the future, this approach can help to analyze long-term simulations (Dameris et al., 2005) and to attribute changes in ozone e.g. to climate variability patterns. It would also be a reasonable tool to identify model to model differences, since it provides a possibility to separate transport and chemical processes.

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